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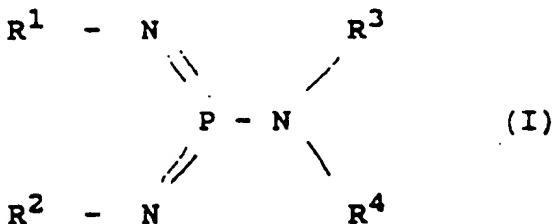
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(54) Polyethylene and process of production thereof.

(57) Polyethylene having from 1 to 60 methyl branches and from 1 to 60 hexyl or higher branches per 1000 carbon atoms, a g-value of from 0.5 to 0.9, a limiting viscosity [η] of from 0.005 to 20.0 dl/g as measured at 140°C in o-dichlorobenzene, and not more than 0.2 of terminal double bond in 1000 carbon atoms, and process for producing the same, comprising polymerising ethylene in the presence of a catalyst composed of a co-ordination nickel compound of bivalent nickel, an organoaluminum compound, and an amino-bis(imino)-phosphorane represented by the general formula (I) below:



where R¹, R², R³ and R⁴ may be the same or different mutually and are respectively n-alkyl, isoalkyl, aryl, or trialkylsilyl.

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POLYETHYLENE AND PROCESS OF PRODUCTION THEREOF

Background of the Invention:

Field of the Invention:

5 The present invention relates to novel polyethylene which has a branching structure. The present invention also relates to a process for the production of such a polyethylene.

Related Background Art:

10 Generally, polymerization of ethylene with a free radical initiator at exceedingly high temperature and pressure gives polyethylene which has long chain branches having length comparable to the main polymer chain thereof. On the contrary, polymerization of ethylene with a Ziegler-Natta catalyst gives polyethylene which has almost no branches.

15 In order to produce branched polyethylene by use of a Ziegler-Natta catalyst, ethylene is generally copolymerized with α -olefin. The copolymerization, however, does not give long chain branches such as those formed by radical polymerization.

20 In an attempt to obtain low-pressure polyethylene having such long chain branching, ethylene may be oligomerized by use of a nickel catalyst such as disclosed in Japanese Patent Application Laid-Open No. 63-12607, and the resulting oligomer may be copolymerized with ethylene in the presence of a chromium catalyst. This method does not give such a long chain branching as produced by free radical polymerization.

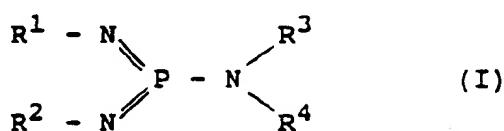
Summary of the Invention:

25 An object of the present invention is to provide a novel polyethylene having a branching structure.

Another object of the present invention is to provide a process for producing the novel polyethylene having branching structure.

30 According to an aspect of the present invention, there is provided polyethylene having from 1 to 60 methyl branches and from 1 to 60 hexyl or higher branches per 1000 carbon atoms, a g-value of from 0.5 to 0.9, and a limiting viscosity $[\eta]$ of from 0.005 to 20.0 dl/g as measured at 140°C in o-dichlorobenzene.

35 According to another aspect of the present invention, there is provided a process for producing polyethylene, comprising polymerizing ethylene in the presence of a catalyst composed of a coordination nickel compound of bivalent nickel, an organoaluminium compound, and an amino-bis(imino)-phosphorane represented by the general formula (I) below:



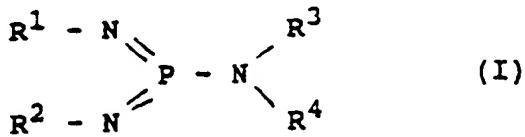
40 where R¹, R², R³ and R⁴ may be the same or different and represent n-alkyl, isoalkyl, aryl, or trialkylsilyl.

The polyethylene of the present invention has from 1 to 60 methyl branches and from 1 to 60 hexyl or higher branches per 1000 carbon atoms as short branches in its structure. The branches are confirmed to exist by ¹³C-NMR. The short branches can be assigned on the basis of J.C. Randall, J. Polym. Sci., Polym. Phys. Ed. 11, 275 (1973), for example.

45 In the structure of polyethylene, the existence of long chain branches may be presumed if the polyethylene has a g-value, namely $g = [\eta]/[\eta]_L$, of 1 or below, where $[\eta]$ is the limiting viscosity of a branched polyethylene, and $[\eta]_L$ is the limiting viscosity of a linear polyethylene having the same melt index. The branched polyethylene 50 of the present invention has a g-value of from 0.5 to 0.9, which suggests the existence of long branches having comparable length to the main polymer chain.

55 The polyethylene of the present invention is completely novel in that it has neither ethyl branches nor butyl branches which exist in polyethylene produced by radical polymerization at high temperature and high pressure. Furthermore, this novel polyethylene has almost no terminal double bonds, which is confirmed by infrared absorption spectrum, the number of double bonds being not more than 0.2 per 1000 carbon atoms. Such polyethylene having no double bonds is characteristically stable during processing.

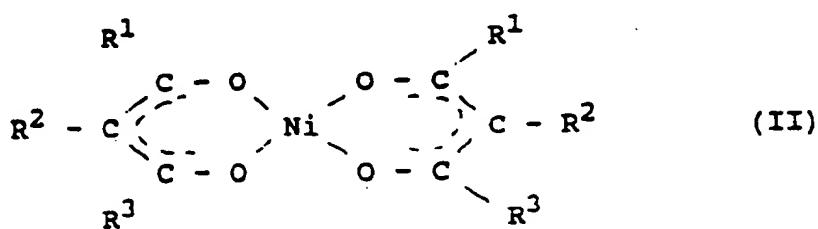
The novel polyethylene can be produced by homopolymerizing ethylene at a temperature of from -78°C to 200°C under an ethylene pressure of from 1 to 200 kg/cm² in the presence of a catalyst composed of a coordination nickel compound of bivalent nickel, an organoaluminium compound, and an amino-bis(imino)-phosphorane represented by the general formula (I) below:



10

where R¹, R², R³, and R⁴ may be the same or different mutually and are respectively n-alkyl, isoalkyl, aryl, or trialkylsilyl.

15 The coordination nickel compound of bivalent nickel includes specifically nickel compounds having β-diketone as ligands, represented by the general formula (II) below:



25

where R¹, R² and R³ may be the same or different and represent hydrogen, n-alkyl (having 1 to 4 carbons), isoalkyl (having 3 to 5 carbons), tert-alkyl (having 4 to 6 carbons), n-fluoroalkyl (having 1 to 4 carbons) or phenyl; nickel stearate; nickel acetate; and the like.

30 The organoaluminium compounds are those represented by the general formula, AlR_sX_{3-s}, wherein s is an integer of 2 or 3; R is alkyl of 1 to 12 carbons; and X is halogen or alkoxy. Specifically the organic aluminium compound may be trimethylaluminium, triethylaluminium, triisopropylaluminium, tri-n-propylaluminium, tri-isobutylaluminium, tri-n-butylaluminium, tri-n-hexylaluminium, tri-n-octylaluminium, tri-n-decylaluminium, tri-n-dodecylaluminium, dimethylaluminium chloride, diethylaluminium chloride, diisopropylaluminium chloride, di-n-propylaluminium chloride, diisobutylaluminium chloride, di-n-butylaluminium chloride, di-n-hexylaluminium chloride, di-n-octylaluminium chloride, di-n-decylaluminium chloride, di-n-dodecylaluminium chloride, dimethylaluminium ethoxide, diethylaluminium ethoxide, di-n-propylaluminium ethoxide, di-n-butylaluminium ethoxide, di-n-hexylaluminium ethoxide, di-n-octylaluminium ethoxide, di-n-decylaluminium ethoxide, di-n-dodecylaluminium ethoxide, and the like.

35 The amino(imino)phosphorane represented by the general formula (I) includes specifically bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane, and the like. Such a compound may be prepared, for example, according to the method of O.J. Scherer and N. Kush: Chem. Ber. 107, 2123 (1974).

40 In the polymerization of ethylene by use of the three-component catalyst, the ratio (molar ratio) of the nickel compound to the amino-bis(imino)phosphorane is preferably in the range of from 1:1 to 1:100. The ratio (molar ratio) of the nickel compound to the aluminium compound is preferably in the range of from 1:1 to 1:1000. The respective components may be introduced either in a solid form or in a solution in a solvent into a polymerization vessel. In adding the catalyst components, preferably the nickel compound and the amino(imino)phosphorane are first brought into contact with each other. Ethylene may be polymerized either in a liquid phase or in a gaseous phase. The polymerization in a liquid phase is conducted preferably by use of an inert solvent. Any of the inert solvents which are generally used in the related technical field may be used, including aliphatic hydrocarbons, aromatic hydrocarbons, and halogenated hydrocarbons having from 4 to 20 carbons. Specific examples are hexane, heptane, pentane, octane, decane, cyclohexane, benzene, toluene, xylene, chlorobenzene, ethylene dichloride, kerosene, and the like.

45 The polyethylene of the present invention can be produced by polymerization of ethylene at a low pressure. Polyethylene having such a structure has not been produced by a low pressure process.

50 The invention may be put into practice in various ways and a number of specific embodiments will be described to illustrate the invention with reference to the accompanying examples and drawing, in which:

55 Figure 1 shows the ¹³C-NMR spectrum of the polyethylene obtained in Example 1.

EXAMPLE 1

5 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 1.0 mmol of nickel acetylacetone, and 5.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 8.0 mmol of triisobutylaluminium was added thereto, and the internal temperature was controlled to be at 40°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 61 g of polymer.

10 The melting point was 77.0°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 1.14 dl/g at 140°C in o-dichlorobenzene.

15 Figure 1 shows the ¹³C-NMR spectrum of the resulting polymer. The peaks ascribable to a methyl branch are observed at 20.0 ppm, 27.5 ppm, 30.4 ppm, 33.2 ppm and 37.5 ppm; the peaks ascribable to the hexyl or higher branch are observed at 14.3 ppm, 23.1 ppm, 27.4 ppm, 30.7 ppm, 32.4 ppm, 34.6 ppm, and 38.3 ppm; and no other peak ascribable to branching was observed. From the spectrum, 13 methyl branches and 25 hexyl and higher branches are found to exist per 1000 carbon atoms. The g-value of the polymer was 0.86.

20 An infrared spectrum showed almost no peaks observable at 890 cm⁻¹, 910 cm⁻¹, and 965 cm⁻¹ which are ascribable to a terminal double bond. Thus the terminal double bond was 0.1 or less in number per 1000 carbon atoms.

EXAMPLE 2

25 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of nickel acetylacetone, and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmol of triisobutylaluminium was added thereto, and the internal temperature was controlled to be at 30°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 174 g of polymer.

30 The melting point was 84.6°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 1.83 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 14 per 1000 carbon atoms, and that of hexyl and higher branches was 19 per 1000 carbon atoms according to ¹³C-NMR spectrum. The g-value was 0.71, and the number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 3

35 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of nickel acetylacetone, and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmol of diethylaluminium chloride was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 100 g of polymer.

40 The melting point was 87.2°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 1.54 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 12 per 1000 carbon atoms, and that of hexyl and higher branches was 14 per 1000 carbon atoms according to ¹³C-NMR spectrum. The g-value was 0.72, and the number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 4

45 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of nickel acetylacetone, and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 18.0 mmol of triethylaluminium was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reac-

tion, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 100 g of polymer.

5 The melting point was 92.0°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 1.98 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 19 per 1000 carbon atoms, and that of hexyl and higher branches was 17 per 1000 carbon atoms according to ^{13}C -NMR spectrum. The g-value was 0.66, and the number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 5

10 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of nickel acetylacetone, and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmols of diethylaluminium ethoxide was added thereto, and the internal temperature was controlled to be at 30°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 24 g of polymer.

15 The melting point was 73.0°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 1.12 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 30 per 1000 carbon atoms, and that of hexyl and higher branches was 18 per 1000 carbon atoms according to ^{13}C -NMR spectrum. The g-value was 0.75, and the number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 6

20 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of bis-(1,1,1,5,5-hexafluoro-2,4-pentanediono)-nickel(II), and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmol of triisobutylaluminium was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 4 g of polymer.

25 The melting point was 101°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 0.96 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 12 per 1000 carbon atoms, and that of hexyl and higher branches was 14 per 1000 carbon atoms according to ^{13}C -NMR spectrum. The number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 7

30 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of bis-(1,3-diphenyl-1,3-propanediono)-nickel(II), and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmol of triisobutylaluminium was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 21 g of polymer.

35 The melting point was 93°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 2.88 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 17 per 1000 carbon atoms, and that of hexyl and higher branches was 6 per 1000 carbon atoms according to ^{13}C -NMR spectrum. The g-value was 0.68, and the number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 8

40 Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of bis-(2,6-dimethyl-3,5-heptanediono)-nickel(II), and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmol of triisobutylalumin-

nium was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 140 g of polymer.

The melting point was 91°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 1.32 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 22 per 1000 carbon atoms, and that of hexyl and higher branches was 17 per 1000 carbon atoms according to ¹³C-NMR spectrum. The g-value was 0.71, and the number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 9

Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of bis-(4-phenyl-2,4-butanediono)-nickel(II), and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmol of triisobutylaluminium was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 84 g of polymer.

The melting point was 88°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 2.15 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 22 per 1000 carbon atoms, and that of hexyl and higher branches was 15 per 1000 carbon atoms according to ¹³C-NMR spectrum. The g-value was 0.68, and the number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 10

Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of bis-(3-oxobutanalato)-nickel(II), and 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)-phosphorane were added. Then 16.0 mmol of triisobutylaluminium was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 2.0 g of polymer.

Two melting points of 90°C and 77°C were observed by differential scanning calorimetry (DSC). The limiting viscosity was 0.74 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 18 per 1000 carbon atoms, and that of hexyl and higher branches was 22 per 1000 carbon atoms according to ¹³C-NMR spectrum. The number of terminal double bonds was not more than 0.2 in 1000 carbon atoms.

EXAMPLE 11

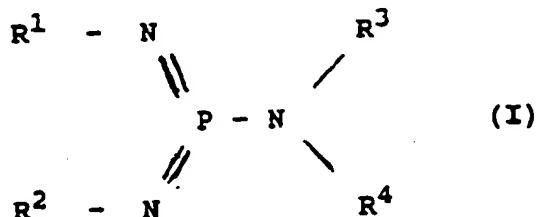
Into a 2-litre stainless steel reactor equipped with a magnetic stirrer, which had been purged thoroughly with nitrogen, 500 ml of toluene, 2.0 mmol of bis-(2-methyl-3-oxobutanalato)-nickel(II), 10.0 mmol of bis(trimethylsilyl)amino-bis(trimethylsilylimino)phosphorane were added. Then 16.0 mmol of triisobutylaluminium was added thereto, and the internal temperature was controlled to be at 20°C. Subsequently, ethylene was introduced therein, and was polymerized for 3.0 hours by keeping the internal pressure at 25 kg/cm²G. After the completion of the reaction, the unreacted ethylene was removed, and the catalyst was decomposed by hydrochloric acid-methanol solution. The reaction mixture was poured into methanol to recover the resulting polymer, which was dried under reduced pressure for 8 hours, yielding 56 g of polymer.

The melting point was 91°C as measured by differential scanning calorimetry (DSC). The limiting viscosity was 0.91 dl/g at 140°C in o-dichlorobenzene. The number of methyl branches was 19 per 1000 carbon atoms, and that of hexyl and higher branches was 23 per 1000 carbon atoms according to ¹³C-NMR spectrum. The g-value was 0.64, and the number of the terminal double bond was not more than 0.2 in 1000 carbon atoms.

As described above, the present invention provides novel polyethylene which is useful for inflation films, injection-moulded articles, blow-moulded articles, extrusion-coating materials, polymer blend materials, and so forth.

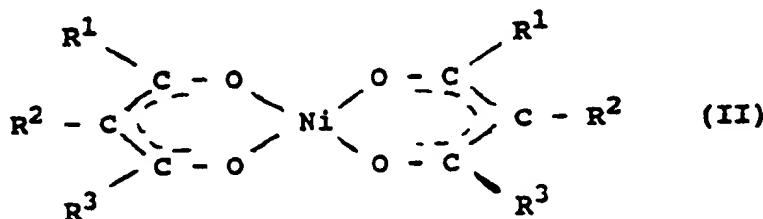
Claims

1. Polyethylene having from 1 to 60 methyl branches and from 1 to 60 hexyl or higher branches per 1000 carbon atoms and a g-value of from 0.5 to 0.9, a limiting viscosity $[\eta]$ of from 0.005 to 20.0 dl/g as measured at 140°C in *o*-dichlorobenzene, and not more than 0.2 terminal double bonds per 1000 carbon atoms.
- 5
2. A process for producing polyethylene, comprising polymerising ethylene in the presence of a catalyst characterised in that the catalyst comprises a co-ordination nickel compound of bivalent nickel, an organoaluminium compound, and an amino-bis(imino)-phosphorane represented by the general formula
- 10 (I) below:



20 where R¹, R², R³ and R⁴ may be the same or different, and each represent an n-alkyl, isoalkyl, aryl or trialkylsilyl group.

- 25 3. A process as claimed in Claim 2 characterised in that the organoaluminium compound is of the general formula AlR_sX_{3-s} , wherein s is an integer of 2 or 3; represents an alkyl group of 1 to 12 carbons; and X represents a halogen atom or an alkoxy group.
4. A process as claimed in Claim 2 or Claim 3 characterised in that the co-ordination nickel compound is a
- 30 nickel compound having -diketone as ligands, represented by the general formula (II) below:



40 wherein R¹, R² and R³ are the same or different and represent a hydrogen atom, an n-alkyl group (having 1 to 4 carbon atoms), an isoalkyl group (having 3 to 5 carbon atoms), a tert-alkyl group (having 4 to 6 carbon atoms), an n-fluoroalkyl group (having 1 to 4 carbon atoms) or a phenyl group.

- 45 5. A process as claimed in Claims 2, 3 or 4 characterised in that the co-ordination nickel compound is nickel stearate or nickel acetate.

FIG. 1

